Geophysical Research Abstracts
Vol. 16, EGU2014-8506, 2014
EGU General Assembly 2014

Towards the simulation of size-resolved aerosol over Europe: model and observations

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Knowledge of the number density and size distribution of atmospheric aerosols in the submicron size range is essential for estimation of the aerosol radiative effects and the health impact of particles. Ultrafine particles (UFP), i.e. particles with diameters $<$100 nm, have received increased attention in recent years with improved possibilities of measuring them. UFP originate from nucleation, involving gas-to-particle conversion, and direct emission from primary combustion processes. As ultrafine particles contribute negligibly to particulate matter mass they should be described in terms of particle number (PN) concentrations. Given the potential health effects due to PN exposure, it is important to obtain PN concentrations with high spatio-temporal resolution. One of the major difficulties in providing accurate estimates of PN is the complexity of atmospheric processes affecting aerosol evolution, e.g. coagulation, deposition, and photochemical formation. Recently, we started the development of the EMEP/MSC-W model towards simulating size-resolved particle number and mass over Europe. Aerosol dynamics in the EMEP model are calculated with the sectional multicomponent aerosol module MAFOR. The performance of MAFOR has been tested extensively against smog chamber experiments and observation data from field campaigns in remote marine and urban environments. The aerosol dynamic processes included are particle nucleation (activation or kinetic parameterisations), coagulation by Brownian motion of particles, and growth due to condensation of sulphuric acid and low volatile/semi-volatile organic vapours (presently only from biogenic VOCs). We present calculations of particle size distributions for 2008 and its geographical and seasonal variation in Europe. As correct modelling of new particle formation (NPF) is essential for accurate predictions of PN concentration, we look closer at model's ability to reproduce several observed NPF episodes. For those NPF episodes, condensational growth, coagulation sink, survival probability and nucleation frequency distribution were additionally analysed. We discuss the sensitivity of calculated particle formation and growth to selected mechanisms and efficiency of nucleation, the saturation pressure of biogenic VOC and size distribution of pre-existing particles. The modelling results show in general a reasonable agreement with median measured PN size distributions in different locations across Europe. The reasons for main discrepancies between the model and observations will be discussed. For instance, secondary particle formation in summer is difficult to capture; the largest identified model deficiency is predicting too frequent nucleation events and too slow growth of 1-10 nm particles at some sites. Possible reasons for this include uncertainties in nucleation parameterizations and BVOC emissions, besides vapours from the photo-oxidation of anthropogenic VOC, which are not yet accounted for. Envisaged modifications of nucleation parameterisation, secondary organic aerosol formation, the implementation of size-resolved ammonium nitrate formation and particle number emissions are expected to further improve model results.